# Correlations of spin currents through a quantum dot induced by the Kondo effect

M. Kindermann[1]

[1] Department of Physics, Massachusetts Institute of Technology, Cambridge MA 02139, USA

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We study correlations of spin currents flowing through a Coulomb blockaded quantum dot. While vanishing for elastic co-tunneling, these correlations develop as the quantum dot enters the Kondo regime. They are a manifestation of Kondo physics in quantum dots. We demonstrate that the spin current correlator is non-perturbative in the Kondo coupling.

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Measurements of current correlations in electrical conductors give valuable information about the microscopic interactions. An important example are shot noise measurements on conductors in the fractional quantum Hall regime [1]. They have demonstrated that the excitations of those interacting systems carry fractions of the elementary charge. Another well-studied interaction phenomenon in nanostructures is the Kondo effect [2]. It is usually observed as an increase of the conductance through a Coulomb blockaded quantum dot (QD) upon lowering its temperature [3]. Its microscopic origin are ground-state correlations between an unpaired spin on the QD and the spins of electrons in the adjacent leads. This suggests that a natural way to reveal Kondo correlations should be through spin-dependent quantities. In this letter we show that indeed Kondo physics manifests itself in correlations of spin currents through a QD. The measurement of spin currents is one of the goals of the rapidly developing field of spintronics [4]. The application discussed here requires a measurement method that does not disrupt the Kondo effect. In particular it must not spin-polarize the leads. One possibility based on the coupling of moving spins to an electric field has been put forward in [5].

We study the correlator

$$C_{\uparrow\downarrow} = \int dt \, \langle I_\uparrow(0) I_\downarrow(t)\rangle - \langle I_\uparrow\rangle\langle I_\downarrow\rangle \tag{1}$$

between the currents $I_\uparrow$ and $I_\downarrow$ of the numbers of spin-up and spin-down electrons flowing through a QD. We assume that the applied bias voltage $V$ and the temperature $T$ are well below level spacing $\epsilon_d$ and charging energy $U$ of the dot, $eV, kT \ll \epsilon_d, U$ (we set $\hbar = k = 1$). The system is then well described by the Anderson single-level impurity model

$$\begin{aligned} H_A &= H_L^{(0)} + H_D + U n_{d\uparrow} n_{d\downarrow} + \sum_{k\beta\sigma} v_\beta (d_\sigma^\dagger c_{k\beta\sigma} + h.c.) \\ H_L^{(0)} &= \sum_{k=-\Lambda^{(0)}_{\beta,\sigma}}^{\Lambda^{(0)}} (\epsilon_k - \mu_\beta) c^\dagger_{k\beta\sigma} c_{k\beta\sigma}, \;\; H_D = -\sum_\sigma \epsilon_d n_{d\sigma}. \end{aligned} \tag{2}$$

$k$ labels the electrons' momentum, $\sigma \in \{\uparrow,\downarrow\}$ their spin and $\beta \in \{L, R\}$ the lead (left or right of the QD), $n_{d\sigma} = d^\dagger_\sigma d_\sigma$. $\mu_\beta$ is the electrochemical potential of lead $\beta$. We have $\mu_L - \mu_R = eV$ and we choose the zero of energy at the Fermi level. If the dot is weakly coupled to the leads, $\Gamma_\beta \ll U - \epsilon_d, \epsilon_d$ ($\Gamma_\beta = 2\pi\nu|v_\beta|^2$ and $\nu$ is the density of states in the leads), and the QD is operated in a Coulomb blockade valley, charge fluctuations on the dot level $d$ are strongly suppressed. If additionally $d$ is occupied by a single electron, the spin of that electron is the only relevant degree of freedom of the QD. One obtains the corresponding low-energy Hamiltonian by a Schrieffer-Wolff transformation [6],

$$\begin{aligned} H^{(0)} &= H_L^{(0)} + H_K^{(0)}, \\ H_K^{(0)} &= \sum_{\substack{k,\beta,\sigma \\ \tilde{k},\tilde{\beta},\tilde{\sigma}}} c^\dagger_{k\beta\sigma} c_{\tilde{k}\tilde{\beta}\tilde{\sigma}} \left( J^{(0)}_{\beta,\tilde{\beta}} \hat{S}_a s^a_{\sigma\tilde{\sigma}} + \frac{\tilde{J}^{(0)}_{\beta,\tilde{\beta}}}{4} \delta_{\sigma\tilde{\sigma}} \right). \end{aligned} \tag{3}$$

$J^{(0)}$ are the amplitudes of scattering processes of lead electrons that involve the spin $S$ of the QD, whereas $\tilde{J}^{(0)}$ accounts for regular potential scattering. $s^a = \sigma^a/2$, where $\sigma^a$ are Pauli matrices. We assume that either $\epsilon_d \ll U - \epsilon_d$ or $\epsilon_d \gg U - \epsilon_d$, such that $|\tilde{J}^{(0)}| = |J^{(0)}|$. The relative sign depends on whether transport occurs by emptying the level $d$, that is $\epsilon_d \ll U - \epsilon_d$ ($\tilde{J}^{(0)} = J^{(0)}$), or by doubly occupying it, that is $\epsilon_d \gg U - \epsilon_d$ ($\tilde{J}^{(0)} = -J^{(0)}$).

Before giving the details of the calculation of $C_{\uparrow\downarrow}$ we motivate its results. For this we focus on the case $\tilde{J}^{(0)} = -J^{(0)}$. In the co-tunneling regime, when only processes to lowest order in $J$ are relevant, transport occurs then through virtual states with a doubly occupied level $d$. These virtual states decay into states with one electron of either spin on the QD, contributing to $I_\uparrow$ or $I_\downarrow$ with equal probabilities. To lowest order in $J$ there are therefore no correlations between $I_\uparrow$ and $I_\downarrow$, $C_{\uparrow\downarrow} = 0$. It is important to note, that if $S = \sigma$ co-tunneling does not allow for the transfer of an electron with spin $\sigma$ without flipping $S$ because of the Pauli principle. This changes at low temperatures, in the Kondo regime. Higher order tunneling processes can then transfer electrons with either spin without flipping $S$. Being equally likely, these "non-spin-flip" processes do not contribute to $C_{\uparrow\downarrow}$. The spin current produced by processes that flip $S$, however, is correlated with the state of $S$. Whenever the spin on the dot is "up", it can flip down and make a contribution to $I_\uparrow$. If $S$ is "down", a spin-flip process produces a

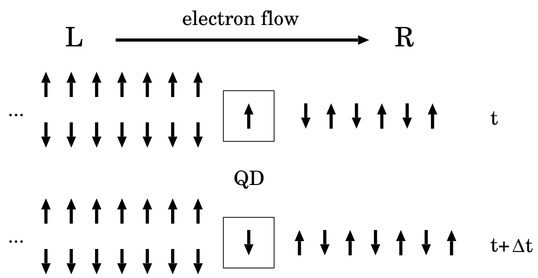


FIG. 1: Illustration of the mechanism that introduces spin-current correlations in the Kondo regime. Only the relevant spin-flip scattering processes are depicted. The QD has different spin at consecutive scattering times $t$ and $t+\triangle t$. Consequently, the outgoing electrons carry alternating spin.

spin-down current. Since in the absence of external spin-relaxation processes every flipping of $S$ to “up” has to be followed by a spin-flip to “down”, both processes occur alternatingly and equally often, as illustrated in Fig. 1. This introduces correlations between $I_\uparrow$ and $I_\downarrow$ and we expect $C_{\uparrow\downarrow}$ to be non-vanishing in the Kondo regime. Formally, the Pauli blocking in the co-tunneling regime is described by Hamiltonian (3) through an interference of the amplitudes $J^{(0)}$ and $\tilde{J}^{(0)}$. It is lifted in the Kondo regime that is described by an effective Hamiltonian of the form (3) with $J^{(0)} \gg \tilde{J}^{(0)}$.

We study the limit of a weak Kondo effect with small effective low-energy couplings $\nu J \ll 1$. One might expect that perturbation theory in $J$ is then adequate to calculate $C_{\uparrow\downarrow}$. This perturbation expansion is, however, plagued by divergences due to the fact that the correlation functions of $S$ do not decay in time to low order in $J$. This indicates that long-time spin correlations have to be treated non-perturbatively. For this we use a method that is close in spirit to the master equation approach to current correlations in sequential tunneling by Bagrets and Nazarov [7]. Since we are interested in the co-tunneling and the Kondo regime we cannot directly apply their method and we derive a variant of it. Our approach is quantum mechanical at short time scales while non-perturbative and classical at times that exceed the decoherence time of $S$.

In the limit of interest $\nu J \ll 1$, we can apply the perturbative renormalization group [8] to capture the short-time spin dynamics. In [9] this approach has been extended to non-equilibrium situations as considered here. Following [9] we eliminate virtual transitions into high energy electron states in the leading logarithm approximation down to a scale $\Omega \gtrsim eV$. This results in an effective Hamiltonian $H$ that has the form of Eq. (3), but renormalized coupling constants $J$ and $\tilde{J}$ instead of $J^{(0)}$ and $\tilde{J}^{(0)}$ and reduced bandwidth $\Lambda$ (instead of $\Lambda^{(0)}$) with $\epsilon(\Lambda) = \Omega$. Spin fluctuations enhance the spin-flip amplitude to $J = 1/2\nu \ln(\Omega/T_K)$, where $T_K$ is the Kondo temperature, while $\tilde{J} = \tilde{J}^{(0)}$. Our limit $\nu J \ll 1$ implies $eV \gg T_K$. We additionally assume that the temperature is smaller but of the same order of magnitude as the voltage, $eV \gtrsim T$.

The central object in our approach is the generating function of spin-current correlators

$$\mathcal{Z}(\lambda) = \mathrm{Tr}\, e^{-i\lambda_{\beta\sigma}N^{\beta\sigma}}\, e^{-iHt}\, e^{i\;\lambda_{\beta\sigma}N^{\beta\sigma}}\, \rho^{(\mathrm{in})}\, e^{iHt}. \qquad (4)$$

$\rho^{(\mathrm{in})}$ is the initial density matrix of the conductor and $\mathcal{Z}$ generates moments of the number $\triangle N^{\beta\sigma}$ of spin $\sigma$ electrons that is transferred into lead $\beta$ during time $t$,

$$\left\langle \prod_{\beta,\sigma} \left(\triangle N^{\beta\sigma}\right)^{p_{\beta\sigma}} \right\rangle = \mathcal{Z}^{-1} \prod_{\beta,\sigma} \left( i \frac{\partial}{\partial \lambda_{\beta\sigma}} \right)^{p_{\beta\sigma}} \mathcal{Z}(\lambda)\Big|_{\lambda=0}. \qquad (5)$$

$\mathcal{Z}$ as defined in Eq. (4) is a Keldysh partition function. Its two time development operators can be implemented by operators on the two branches of the Keldysh time-contour [10]. The exponentials $\exp\{\pm i\lambda_{\beta\sigma}N^{\beta\sigma}\}$ can be absorbed into phases for the tunneling terms in $H$. By inserting complete sets of Fermion coherent states [11] we then derive a mixed representation [12] of $\mathcal{Z}$ as a path integral over a time-ordered spin operator expression,

$$\mathcal{Z}(\lambda) = \mathrm{Tr}_S\, T_c \int \mathcal{D}c^*\mathcal{D}c\, \rho_S^{(\mathrm{in})}\, e^{-ic^*(\mathcal{G}^{-1}+\mathcal{J}^\lambda)c}. \qquad (6)$$

The Fermion fields $c$ carry Keldysh, lead, spin, momentum, and frequency indices $\alpha$, $\beta$, $\sigma$, $k$, and $\omega$. $\mathcal{G}$, a matrix in this space, is the electron Green function corresponding to $H_L$. $\mathcal{J}^\lambda$ describes tunneling processes and contains spin operators. $T_c$ denotes operator ordering along the Keldysh time-contour and relative to the initial spin density matrix $\rho_S^{(\mathrm{in})}$. The trace $\mathrm{Tr}_S$ over spin operators is taken. We have

$$\mathcal{J}^\lambda_{\alpha\beta\sigma k\omega,\alpha'\beta'\sigma'k'\omega'} = \tau^z_{\alpha\alpha'} \left( J_{\beta\beta'} s^a_{\sigma,\sigma'} \hat{S}^{\alpha,\omega\omega'}_a + \frac{\tilde{J}_{\beta\beta'}}{4} \delta_{\sigma\sigma'}\delta_{\omega\omega'} \right) e^{-i(\lambda_{\beta\sigma}-\lambda_{\beta'\sigma'})\tau^z_{\alpha\alpha'}/2} \qquad (7)$$

with the third Pauli matrix $\tau^z$ and spin operators $\hat{S}^{\alpha,\omega\omega'}_a = \int dt\, e^{-i(\omega-\omega')t}\hat{S}^\alpha_a(t)$ on the Keldysh branch $\alpha$. Integrating over the Fermions in Eq. (6) we obtain

$$\mathcal{Z}(\lambda) = \mathrm{Tr}_S\, T_c\, \rho_S^{(\mathrm{in})}\, e^{\mathrm{Tr}\,\ln(1+\mathcal{G}\mathcal{J}^\lambda)}\, \mathcal{Z}|_{J=\tilde{J}=0} = \mathrm{Tr}_S\, T_c\, \rho_S^{(\mathrm{in})}\, e^{\mathrm{Tr}\,[\mathcal{G}\mathcal{J}^\lambda - \frac{1}{2}\mathcal{G}\mathcal{J}^\lambda\mathcal{G}\mathcal{J}^\lambda + \mathcal{O}(J^3)]} \equiv \mathrm{Tr}_S\, T_c\, \rho_S^{(\mathrm{in})}\, e^{-\mathcal{L}^\lambda}. \qquad (8)$$

The $\mathcal{O}(J)$ term of $\mathcal{L}^\lambda$ does not contain spin operators, while the terms of $\mathcal{O}(J^3)$ are negligible for $\nu J \ll 1$. To save space we evaluate here only the $\mathcal{O}(J^2)$ term $\mathcal{L}_o^\lambda$ that derives from the spin off-diagonal matrix elements of $\mathcal{J}^\lambda$,

$$\mathcal{L}_o^\lambda = \sum_{\substack{k,k'\\ \alpha\alpha'\beta\beta'}} \frac{|J_{\beta\beta'}|^2}{4} \tau^z_{\alpha\alpha}\tau^z_{\alpha'\alpha'} e^{i(\lambda_{\beta\uparrow}-\lambda_{\beta'\downarrow})(\tau^z_{\alpha\alpha}-\tau^z_{\alpha'\alpha'})/2} \int dt\, dt'\, G^{\beta k}_{\alpha,\alpha'}(t'-t) G^{\beta' k'}_{\alpha',\alpha}(t-t') \hat{S}_-^{\alpha'}(t)\hat{S}_+^{\alpha}(t'), \tag{9}$$

where $G^{\beta k}_{\alpha,\alpha'}(t) = \int (d\omega/2\pi)\, e^{-i\omega t}\mathcal{G}_{\alpha\beta\uparrow k\omega,\alpha'\beta\uparrow k\omega}$ and $\hat{S}^\alpha_\pm = \hat{S}^\alpha_x \pm i\hat{S}^\alpha_y$. The Green functions in Eq. (9) summed over $k, k'$ decay exponentially over the time $\tau_T = 1/T$. At time scales longer than $\tau_T$ $\mathcal{L}_o^\lambda$ is therefore local in time. It moreover couples spin-flip processes occurring at the same time on different branches of the Keldysh contour. This leads to classical behavior at long times and it allows for a description along the lines of [7]. One could obtain the effective theory on the scale $\tau_T$ by integrating out fast spin fluctuations in a path integral for the original model Eq. (3) [13]. Equivalently we integrated out high frequency spin fluctuations in the Hamiltonian formalism. Due to the form of the Green functions in the effective model $H$ with reduced bandwidth, $\mathcal{L}^\lambda$ is then strongly suppressed for frequencies $\omega > \Omega$ and these fluctuations contribute to $\mathcal{Z}$ only negligibly. The corrections to the scaling logarithms due to spin fluctuations in the frequency range $T < \omega < \Omega$ are for our choice of parameters $\Omega \gtrsim eV \gtrsim T$ of $\mathcal{O}(1)$ and to leading order in the logarithms negligible as well. To this accuracy $\mathcal{L}_o^\lambda$ therefore equals the corresponding piece of the effective theory at the scale $\tau_T$. The contribution due to the diagonal elements of $\mathcal{J}^\lambda$ has an analogous structure and we conclude that $\mathcal{L}^\lambda$ is local on the time scale $\tau_T$. In Eq. (8) we have expressed $\mathcal{Z}$ as the trace over a time dependent density matrix $\rho_S^\lambda(t) = T_c\, \rho_S^{(\mathrm{in})}\, \exp(-\mathcal{L}^\lambda)$. Because of the locality of $\mathcal{L}^\lambda$, $\rho_S^\lambda$ obeys an ordinary differential equation. The off-diagonal entries of $\rho_S^\lambda$ decay exponentially under evolution with that equation. This shows that the spin dynamics is indeed classical on the time scale $\tau_T$. It is therefore sufficient to study the evolution of the diagonal elements $p_\uparrow^\lambda$ and $p_\downarrow^\lambda$ of $\rho_S^\lambda$, that we collect into a vector $p^\lambda = (p_\uparrow^\lambda, p_\downarrow^\lambda)$. $p^\lambda$ obeys

$$\partial_t p^\lambda = -\hat{L}^\lambda p^\lambda, \tag{10}$$

$$\hat{L}^\lambda = \begin{pmatrix} \Gamma_S - \sum_{\beta\beta'} a_{\beta\beta'} \left( \left|\frac{J+\tilde{J}}{2}\right|^2 C^\lambda_{\beta\uparrow,\beta'\uparrow} + \left|\frac{J-\tilde{J}}{2}\right|^2 C^\lambda_{\beta\downarrow,\beta'\downarrow}\right) & -\Gamma_S - \sum_{\beta\beta'} a_{\beta\beta'}|J|^2 C^\lambda_{\beta\downarrow,\beta'\uparrow} \\ -\Gamma_S - \sum_{\beta\beta'} a_{\beta\beta'}|J|^2 C^\lambda_{\beta\uparrow,\beta'\downarrow} & \Gamma_S - \sum_{\beta\beta'} a_{\beta\beta'} \left( \left|\frac{J+\tilde{J}}{2}\right|^2 C^\lambda_{\beta\downarrow,\beta'\downarrow} + \left|\frac{J-\tilde{J}}{2}\right|^2 C^\lambda_{\beta\uparrow,\beta'\uparrow}\right) \end{pmatrix}, \tag{11}$$

$$\begin{aligned} C^\lambda_{\beta\sigma,\beta'\sigma'} &= e^{-i(\lambda_{\beta\sigma}-\lambda_{\beta'\sigma'})} - 1, \\ a_{\beta\beta'} &= (\pi\nu)^2 \int \frac{d\epsilon}{2\pi}[1-f_\beta(\epsilon)]f_{\beta'}(\epsilon). \end{aligned} \tag{12}$$

For conciseness we assume from now on that the QD is symmetrically coupled to the leads, $J_{\beta\beta'} = J$, $\tilde{J}_{\beta\beta'} = \tilde{J}$. $a_{\beta\beta'}|J|^2$ are the usual rates for tunneling between two leads. $\Gamma_S = \sum_{\beta\beta'} a_{\beta\beta'}|J|^2 + \gamma_S$ is the relaxation rate of $S$. The first term accounts for spin-flips by conduction electrons. The second term $\gamma_S$ has been introduced phenomenologically. It accounts for spin-flip processes that are not described by our model. We assume $\gamma_S \ll T$, such that this additional spin-decoherence does not affect Kondo correlations. Eqs. (10) with (11) differ from their counterparts for sequential tunneling [7] mainly in how the electron state occupation numbers enter. Kondo correlations are accounted for by the renormalization of $J$. The off-diagonal entries of $\hat{L}^\lambda$ describe spin-flips of $S$. Because electrons can also be transferred without flipping $S$, the diagonal elements of $\hat{L}^\lambda$ contain counting factors $C^\lambda$ as well.

We integrate Eq. (10) to obtain $\mathcal{Z}$. At long times $t$ it simplifies to the exponential of the smallest eigenvalue of $-t\hat{L}^\lambda$. $C_{\uparrow\downarrow}$ can then be obtained using Eq. (5). We introduce transmission probabilities $\tau = (2\pi\nu J)^2$, $\tilde{\tau} = (2\pi\nu\tilde{J})^2$ and the spin-off-diagonal Fano-factor $F_{\uparrow\downarrow} = C_{\uparrow\downarrow}/I$ ($I$ is the mean current through the QD). Although not within the limits of applicability of our theory, it is instructive to first take the zero temperature limit

$$F_{\uparrow\downarrow} = \frac{eV\tau}{2(3\tau+\tilde{\tau})(eV\tau + 8\pi\gamma_S)}\,(\tau - \tilde{\tau}). \tag{13}$$

Eq. (13) displays most clearly the main finding of this letter: While $F_{\uparrow\downarrow}$ vanishes in the absence of Kondo correlations, when potential and spin-flip scattering processes have equal probabilities $\tau = \tilde{\tau}$, it grows non-zero in the Kondo regime. We need to convince ourselves that this carries over to finite temperature $T \simeq eV$.

The first correction to Eq. (13) due to temperature, $\triangle F^{(1)}_{\uparrow\downarrow} = [(\tilde{\tau}-\tau)/(3\tau+\tilde{\tau})](T/eV)$ ($\gamma_S = 0$) is reassuring: it again vanishes unless there are Kondo correlations. The full temperature dependence of $F_{\uparrow\downarrow}$,

$$F_{\uparrow\downarrow} = \frac{\tau v^2[\tau\cosh^2(v/2) - \tilde{\tau}\sinh^2(v/2)] - 4\tau(\tau + 8\pi\gamma_S/T)\sinh^2(v/2)}{(3\tau+\tilde{\tau})v[\tau v\sinh v + 4(\tau + 4\pi\gamma_S/T)\sinh^2(v/2)]}, \quad v = \frac{eV}{T}, \tag{14}$$

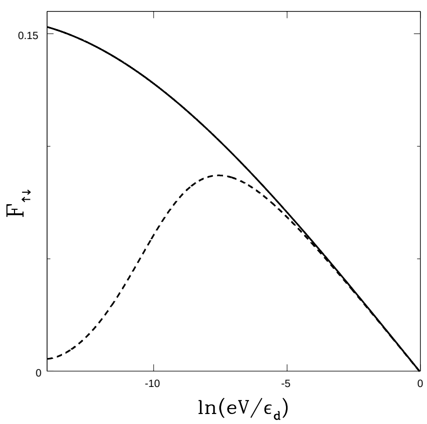


FIG. 2: Voltage dependence of the Fano factor $F_{\uparrow\downarrow}$ for $T = eV/30$, $\epsilon_d = 1\,\text{meV}$, and $T_K = 10^{-7}\,\epsilon_d$ (solid line: $\gamma_S = 0$, dashed line: $\gamma_S = 10^5\,\text{s}^{-1}$). $F_{\uparrow\downarrow}$ develops in parallel with Kondo correlations upon lowering the voltage/temperature.

is shown in Fig. 2. In this plot both temperature and voltage are varied, at constant ratio $eV/T \gtrsim 1$. Although the Fano factor $F_{\uparrow\downarrow}$ at finite temperature does not reach its theoretical zero temperature maximum $1/6$, its qualitative behaviour is robust: it develops in parallel with Kondo correlations. The effect is cut-off at small voltages $eV \ll \gamma_S/\tau$ by external spin-flip processes. $S$ is then flipped randomly in between electron transfers and the mechanism illustrated in Fig. 1 is inoperative. For Fig. 2 we have assumed typical experimental parameters $\epsilon_d = 1\,\text{meV}$ and $\gamma_S = 10^5\,\text{s}^{-1}$. Even longer spin relaxation times have been observed [14].

We come back now to the difficulties encountered with perturbation theory in $J$. The zero temperature limit Eq. (13) shows most clearly that $C_{\uparrow\downarrow}$ in the model Eq. (3) ($\gamma_S = 0$) is non-perturbative in $J$: the limit $\gamma_S \to 0$ implies that $\tau \gg \gamma_S/eV$, that is $J^2 \gg \gamma_S/\nu^2 eV$ and it cannot be accessed in an expansion around $J = 0$. It lies outside its radius of convergence. Perturbation theory in $J$ misses correlations of scattering events over the timescale $1/\Gamma_S$. These long-time correlations are manifest in the frequency spectrum of $C_{\uparrow\downarrow}$. It can be obtained by a straightforward extension of our method to slowly time-dependent $\lambda$. The approach is valid for frequencies $\omega \ll T$ and yields

$$C_{\uparrow\downarrow}(\omega) = \left(IF_{\uparrow\downarrow} + \frac{\tau T}{8\pi}\right)\frac{\Gamma_S^2}{\omega^2/4 + \Gamma_S^2} - \frac{\tau T}{8\pi}. \tag{15}$$

The dispersion of $C_{\uparrow\downarrow}$ on the scale $\Gamma_S$, that is of the order of the mean current $I/e$ at small $\gamma_S$, is a consequence of the time-ordering of spin-flips described above.

In conclusion, we have studied correlations of spin currents through a QD. We have found that these correlations can be induced by Kondo fluctuations on the QD. They are a new manifestation of the Kondo effect in QDs.

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